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## Measurement of isotope shift in Eu II

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### Abstract

The isotope shift between singly-charged  $^{151}\text{Eu}$  and  $^{153}\text{Eu}$  in the  $4f^7(^8S^o)6s\ ^9S_4^o / - - 4f^7(^8S^o)6p_{1/2}\ J = 4$  transition at 4129 ] has been measured using fast ion beam-laser technique. This Eu line has attracted interest in connection with efforts of obtaining a cosmochronometer based on observed Th/Eu abundance ratios. Knowledge of the isotope shift is of importance in order to check that contaminations from line blends do not contribute to the line intensity of Eu II. The measured value of the isotope shift  $-0.1527(2)\text{cm}^{-1}$  is consistent with the old spectroscopic value of Krebs and Winkler  $(-0.1503(25)\text{cm}^{-1})$  using a Fabry Perot interferometer, while the accuracy is improved substantially.

## 1. Introduction

Stellar spectra can be used for determination of elemental abundances. Recently Butcher [1] proposed that stellar spectra also could be used for age determination of stars. The only long-lived isotope of thorium,  $^{232}\text{Th}$ , has a half-life of 14 Gyr, which relates well to the time scale for stellar and galactic evolution. A useful cosmochronometer might be obtained if the thorium abundance is compared to a stable element [1]. Singly-charged thorium (Th II) has a useful spectral line at 4019.129 Å. Butcher [1] chose a nearby transition in singly ionized neodymium (Nd II) at 4018.823 Å for comparison. He extracted abundance ratios of these spectral lines for stars of very different ages and concluded that the galactic age is no more than at maximum 9.6 Gyr, if a constant rate of nucleo-synthesis throughout of the life of the Galaxy was assumed. For an exponentially decaying production rate he obtained 11-12 Gyr.

The proposed method involves some problems regarding both Th and Nd. The Th II line is blended with a Co I line [2]. The cobalt line is much more sensitive to temperature which causes the contribution from cobalt to vary very much from star to star. Lawler et al [2] have analyzed the contamination from cobalt in the Th II intensities of the same set of stars that was analyzed by Butcher. From the revised analysis of the spectra and in comparison with predictions based on models of chemical evolution, they suggested an age of the Galaxy of 15-20 Gyr. A more detailed analysis, utilizing spectrum synthesis, by Morell et al [3] revealed the need to introduce further blends in the spectrum close to the position of the Th II line in order to reproduce the spectrum. They conclude that due to the scattering in the Th/Nd abundance ratio, it is not possible to draw any firm conclusion about the age of the Galaxy. A further complication regarding the Th II line was discussed recently by Grevesse and Noels [4]. They found that the solar Th abundance derived from the 4019 Å line intensity was 40% that this discrepancy could be explained and corrected for by a blend of the V I intercombination line  $a\ ^2D_{5/2} - w\ ^4P_{5/2}$ .

There are also problems with the use of Nd as a stable element reference. The thorium isotope is only created by the r-process (rapid neutron capture), whereas neodymium only partly is formed by this process, the rest being created by the s-process (slow neutron capture). Although Butcher [1] claims that the partial contributions do not vary substantially from star to star, an alternative stable element has been searched for. As suggested by Pagel [5], europium is a good candidate, almost entirely formed by the r-process. A

Eu II line at 4129.70 ] seems to be a good trace of stellar europium abundance. The line appears to be free from line blends (and its wavelength is not too far from that of the Th II line). Thus, it has been suggested that this line should be compared with the Th II intensity. Recently, François et al [6] have investigated the Th/Eu ratios in a set of metal poor stars. They obtained a complex variation in the actual ratio, from which no conclusions could be drawn regarding the age of the Galaxy. It is not clear from this paper on which atomic data the analysis was relying. We presume that the Eu II line mentioned here (4129.7 ] ) was used by François et al and that the line 4013 ] mentioned in their article was a misprint. It was not mentioned whether hyperfine structure and isotope shift were considered in the analysis.

It is important that the 4129.7 ] Eu II line in the solar spectrum, with known hyperfine structure and isotope shift for the europium isotopes, can be reproduced otherwise blends must be searched for. A weak blend in the solar spectrum might be strong in other types of stars, especially if the blend comes from a neutral element of the iron group. For this reason it is of great importance to have accurate values for hyperfine structure and isotope shift in order to be able to synthesize a very accurate line profile from which it could be concluded whether or not a blend is present.

The hyperfine structure of the Eu II  $4f^7(^8S^o)6s\ ^9S_4^o - 4f^7(^8S^o)6p_{1/2}\ J = 4$  transition at 4129.70 ] has recently been accurately determined in the two isotopes  $^{151}\text{Eu}$  and  $^{153}\text{Eu}$ . Becker et al [7] have measured the hyperfine splitting of the ground state ( $4f^7(^8S^o)6s\ ^9S_4^o$ ) using laser rf double-resonance technique in a Paul trap. By this method they have been able to determine the hyperfine splittings, which are in the range 1-10 GHz, with an accuracy of the order of 100 Hz. The hyperfine structure of the upper level has been determined by Villemoes and Wang [8] using the fast ion beam-laser method. Here the splittings are of the order 0.1-1 GHz, and these have been determined with an accuracy of about 1 MHz. Thus, there are reliable hyperfine data which can be used in the analysis of stellar spectra.

For the isotope shift there is an old spectroscopic measurement by Krebs and Winkler [9] using a Fabry-Perot interferometer, which partially could resolve the hyperfine components of the spectrum. Krebs and Winkler obtained an isotope shift of  $-0.1503 \pm 0.0025\text{ cm}^{-1}$  for this particular transition. However, due to the efforts of finding a cosmochronometer there has been a request from astrophysicists [10] to remeasure this isotope shift with the accurate beam-laser technique in order to check the old measurement and to firmly

establish the isotope shift for this transition. Thus we decided to perform a measurement of this quantity.

## 2. Experimental set-up

At the CRYRING facility of the Manne Siegbahn Laboratory of Stockholm University there is an ion source CRYISIS which has an ion injector (INIS), which apart from its function as injector also can be used separately for fast ion beam-laser spectroscopy. INIS is an isotope separator usually operated at 25 kV. The experimental set-up has been described in some detail previously [11]. The ions were produced in a low-voltage electron-impact ion source. Europium was introduced in metallic form and the arc in the ion source was supported by letting a noble gas into the discharge region. The two isotopes of europium ( $^{151}\text{Eu}$  and  $^{153}\text{Eu}$ ) are easily separated by the mass analyzing magnet.

An ion current of a few hundred nA was collected for each isotope (the two isotopes have almost the same abundance) in a Faraday cup, which can be inserted in front of the interaction chamber. With a system of electrostatic deflection plates and quadrupoles the beam could be transported into the interaction region.

The laser light was introduced into the vacuum system in antiparallel direction with respect to the ion beam (Fig 1). To assure that the ion beam and the laser beam travel along the same path in the interaction chamber, 4 mm diameter apertures are mounted on each side of the interaction chamber (at a distance of 1.4 m from each other). Inside the chamber the ion beam velocity is locally changed by a Doppler Tuning Device (DTD), consisting of three electrically isolated cylindrical tubes. The center tube is connected to high voltage while the others are grounded. The DTD was located in the focus of the optical fluorescence detecting system. Thus, due to the local change of the Doppler shift, the laser excitation could be restricted to the region where fluorescence could be detected. This is essential in order to avoid reduction of signal intensity by optical pumping of the ion beam upstreams the detection region.

Through a slot in the center tube a lens system collected fluorescence light and focused it on the entrance slit of a 0.35 m Heath EUE-700 monochromator (Czerny-Turner mounting, f/6.8). With slits set at 2 mm, the bandwidth was 40 Å. Photons were counted by a EMI 9789 QA photomultiplier, which was cooled by a Hamamatsu Peltier cooler. More details about the experimental set-up and also the use of the DTD for lifetime measurements can be found in our previous papers [11-13].

A single-mode Coherent 699-29 Autoscan dye laser (with Stilbene 1) was used to provide tunable laser

light at 4129.7 ] (in the rest frame of the ion) with a bandwidth of less than 1 MHz. The ring dye laser was pumped by the UV lines of an Innova 400-20 argon ion laser. About 100 mW tunable single frequency blue laser light was obtained. This laser power, however, turned out to be too low to support the Autoscan function of the ring dye laser. The laser was instead operated in the 699-21 mode, i.e. utilizing the 30 GHz continuous scan option. Since the laser power was too low (in combination with the high laser frequency) to permit use of the internal wavemeter, an external Burleigh WA-4500 wavemeter was used to find the initial laser frequency of the 30 GHz scan. The wavemeter has an accuracy of  $0.02 \text{ cm}^{-1}$ . During the laser scan the wavemeter was monitored in order to observe whether mode hops occurred.

### 3. Experiment

The goal of the present experiment was to measure the isotope shift in the  $4f^7(^8S^o)6s\ ^9S_4^o / - 4f^7(^8S^o)6p_{1/2}\ J = 4$  transition for reasons mentioned above. This transition occurs at 4129.70 ], i.e. 24207.86 [14]  $\text{cm}^{-1}$ . For excitation in antiparallel geometry, the wavenumber of the exciting laser light has to be reduced by about  $15\ \text{cm}^{-1}$  to compensate for the Doppler shift. Fluorescence was observed by monitoring the  $4f^7(^8S^o)6s\ ^7S_3^o - 4f^7(^8S^o)6p_{1/2}\ J = 4$  transition at 4435.55 ] ( $22539\ \text{cm}^{-1}$ ).

The basic idea of the experiment was to switch isotopes during the laser scan in order to relate the hyperfine components of the two isotopes to the same laser frequency scale, without requirements of a very accurate knowledge of the laser frequency on an absolute scale [15]. Consequently the isotope shift will be exposed directly in the spectrum. In the analysis, however, the different Doppler shifts of the two isotopes must be taken into account. Since both isotopes were accelerated by the same high voltage, the velocities will differ due to the isotopes different masses. This Doppler ‘isotope shift’ is of the same order of magnitude as the real isotope shift.

In Fig 2a simulated spectra are shown, which are based on the hyperfine splittings given in refs [7] and [8] (with a resolution similar to that of the present experiment). The simulated spectrum of  $^{151}\text{Eu}$  is displaced relative that of  $^{153}\text{Eu}$  due to the isotope shift as well as due to the different Doppler shifts as indicated schematically in the figure. Each spectrum reflects the different splittings of the upper and the lower level. The large splittings between the lower levels give rise to the large distances between six groups of levels according to Becker et al [7]. The distance between lines inside such a spectral line group reflects the hyperfine splitting of the upper level, as given by Villemoes and Wang [8]. In the present experiment a few survey spectra were recorded to identify the different components of the spectra. In Fig 2b such an example is given.

For the isotope shift measurement we decided to record one three-line group of hyperfine components in  $^{151}\text{Eu}$  ( $\frac{11}{2} - \frac{9}{2}, \frac{11}{2} - \frac{11}{2}, \frac{11}{2} - \frac{13}{2}$ ) (where  $F - F'$  denotes the hyperfine quantum number for the lower and the upper level, respectively), then switch isotope to  $^{153}\text{Eu}$  and record the three-line group ( $\frac{9}{2} - \frac{7}{2}, \frac{9}{2} - \frac{9}{2}, \frac{9}{2} - \frac{11}{2}$ ) and then again change back to isotope  $^{151}\text{Eu}$  and record the three-line group ( $\frac{9}{2} - \frac{7}{2}, \frac{9}{2} - \frac{9}{2}, \frac{9}{2} - \frac{11}{2}$ ); all these nine lines recorded in one single laser scan. The isotope separator was operated by a computer based

control system. This system permitted programming of the magnet steering so that isotopes could be shifted by a single strike of a key (the isotopes shifted in about one second). Several spectra were recorded by this method.

Since the frequencies of the hyperfine splittings are well-known, the spectra could be internally calibrated. The six  $^{151}\text{Eu}$  lines were used for this purpose. The frequency scale was fitted to a third-degree polynomial. As mentioned above the external wavemeter was only used to determine the frequency of the starting point of the laser scan and to check that no mode hops occurred during the scan. The fluorescence photon pulses were amplified, discriminated and read into a CAMAC scaler. A PC was used to read the scaler and store the value in a multi-scaler program as the laser frequency was scanned. A typical spectrum (Fig 3) was recorded in 500 s (1 second per point).

The spectral line positions were determined by a peak-fitting program. To obtain as high intensity as possible in the spectrum, fluorescence was observed close to the field gap of the DTD. This caused the lines to be slightly asymmetric, since in the high frequency wing of the line, fluorescence from excitation in the gap will contribute. In the present experiment the voltage on the DTD was -401 V, yielding a higher beam velocity inside the DTD than outside. In the fitting program the lines were approximated by a Gaussian line profile with an exponential foot on the high frequency wing (to take care of the asymmetry). The Gaussian width reflects the velocity spread in the ion beam. In the present experiment a typical Gaussian width of about 180 MHz (FWHM) was obtained. To obtain the isotope shifts, the experimental hyperfine constants [7,8] were used to determine the centre-of-gravity of the multiplets. The weighted average of the ‘apparent’ isotope shift (i.e. the shift in the recorded spectrum without Doppler correction) could be determined with an accuracy of 4 MHz, where statistical errors both from the calibration procedure and the determination of the peak positions are included.

After the line positions were determined, correction for the different Doppler shifts of the two isotopes had to be done. The beam velocity can be accurately determined if the ion beam is excited both in parallel and antiparallel geometry [16] according to the formula

$$\beta = \frac{v}{c} = \frac{\sigma_+ - \sigma_-}{\sigma_+ + \sigma_-}$$

, where  $\sigma_+$  is the laser frequency for excitation in parallel configuration and  $\sigma_-$  for antiparallel. However, due to the practical circumstances, it was more reliable to apply this technique to a Xe II transition, for which the laser dye Rhodamine 6G could be used. The laser system could thus be run in a more stable mode utilizing the Autoscan option and the use of the internal wave meter. The xenon transition is also stronger and the single line appearing in  $^{132}\text{Xe}$  is unambiguous. By this method the high voltage could be accurately be determined, which could be used to calculate the velocity of the europium isotopes, which were accelerated by the same high voltage.

The high voltage calibration was performed by exciting the Xe II transition  $5d\ ^4D_{7/2} - 6p\ ^4P_{5/2}^o$  at 6051.15] (16521.22 cm $^{-1}$ ) [17] (in the ion rest frame) and observing the laser-induced fluorescence at 5292 ] ( $6s\ ^4P_{5/2} - 6p\ ^4P_{5/2}^o$ ), as the laser frequency was tuned. Iodine lines [18] were used as reference lines. Simultaneously, the Vernier etalon signal (of the internal wave meter) as well as the transmission of an external high finesse Fabry-Perot interferometer (2 GHz FSR) were recorded. Statistics of 16 scans for  $\sigma_-$  and 10 scans for  $\sigma_+$  were used in this calibration procedure and the high voltage was determined to be  $24454 \pm 10$  V. In the calibration procedure the DTD was used in the same position and at the same voltage as in the europium run. Thus it did not matter that the beam was not observed in a completely field free region in the DTD (as found through the calibration measurements). From the calibration of the high voltage the velocities for the europium isotopes could be determined. For -401 V on the DTD  $\beta(^{151}\text{Eu}) = (5.9405 \pm 0.0012) \cdot 10^{-4}$  and  $\beta(^{153}\text{Eu}) = (5.9015 \pm 0.0012) \cdot 10^{-4}$  were found. These  $\beta$ -values were used to correct for the Doppler shifts. Since both isotopes were recorded in the same laser scan, it is only the difference in velocity for the two isotopes which matters which means that this determination is not so critical on an absolute scale. The velocity difference is determined directly from the mass difference, since both isotopes are accelerated by the same high voltage. In the non-relativistic limit  $\beta(^{153}\text{Eu}) = \sqrt{m(^{151}\text{Eu})/m(^{153}\text{Eu})} \cdot \beta(^{151}\text{Eu})$ , which implies that the uncertainty in the isotope shift due to the uncertainty in the difference in velocity ( $\delta(\Delta\beta)$ ) is only  $\Delta\beta[1 - \sqrt{m(^{151}\text{Eu})/m(^{153}\text{Eu})}]\nu \approx 0.5$  MHz (where  $\Delta\beta$  is the uncertainty in  $\beta$  as given above and  $\nu$  is the actual laser frequency). All calculations were, however, performed using the full relativistic expressions for beam energies and Doppler shifts. Also for the internal frequency calibration mentioned above, full relativistic Doppler correction was made. Atomic masses ( $m(^{151}\text{Eu})$  and  $m(^{153}\text{Eu})$ ) were obtained from the



mass tables of Wapstra and Bos [19].

#### 4. Results and discussion

From the evaluation of the measurements described above an isotope shift of  $-152.7 \pm 0.2$  mK (i.e.  $-0.1527$  cm<sup>-1</sup> or 4578 MHz) was obtained. The present result coincides within error bars with the value of Krebs and Winkler [9], who obtained an isotope shift of  $-150.3 \pm 2.5$  mK for this particular transition. It is impressive that the old classical work could give such an accurate result though the different hyperfine components were not very well resolved.

With the present result for the isotope shift and the recent accurate values for the hyperfine structure [7,8], there is now a solid ground of atomic data which can be used in the analysis of europium in stellar spectra.

In the interpretation of stellar spectra more caution might be needed due to the recent observations of isotopic abundance anomalies (see e.g. [20]). The relative intensity between the <sup>151</sup>Eu and the <sup>153</sup>Eu lines might not be a universal constant for different objects.

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## FIGURE CAPTIONS

Fig 1

Schematic figure of the experimental set-up used for the isotope shift measurements in europium.

Fig 2

a. Simulated spectra of  $^{151}\text{Eu}$  and  $^{153}\text{Eu}$  using the hyperfine constants given by Becker et al [7] and Villemoes and Wang [8]. The spectra are shifted relative each other according to the different Doppler shifts at the beam energy used in the present experiment. The figure illustrates how the centres-of-gravity of the multiplets shift due to the isotope shift and the difference in Doppler shift to give the observed shift (which is denoted by the ‘apparent’ isotope shift in the text).

b. An experimental survey spectrum covering 14 of the 16 components of the  $4f^7(^8S^o)6s\ ^9S_4^o / - - 4f^7(^8S^o)6p_{1/2}\ J = 4$  transition in  $^{153}\text{Eu}$ .

Fig 3

One of the experimental spectra used for extracting the isotope shift between  $^{151}\text{Eu}$  and  $^{153}\text{Eu}$  in the  $4f^7(^8S^o)6s\ ^9S_4^o / - - 4f^7(^8S^o)6p_{1/2}\ J = 4$  transition in Eu II. The spectrum shows detected laser-induced fluorescence from the ion beam as the frequency of the ring dye laser was continuously scanned. During the laser scan the isotope in the ion beam was changed. The spectrum starts with fluorescence from  $^{151}\text{Eu}^+$ . When the three first components had been recorded, the setting of the magnet of isotope separator was instantly switched to give  $^{153}\text{Eu}^+$ . Three components were recorded in this isotope. Then the beam was switched back to the  $^{151}\text{Eu}^+$  isotope and three new components were recorded in this isotope. This procedure permits internal calibration of the frequency scale using the accurate hyperfine splitting reported by Becker et al [7].